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Thermochemical Evaluation of Proposed Electrothermal-Chemical Propellants

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Due to the addition of electrical energy, the customary thermodynamic method of screening candidate fuels needs to be modified for application to electrothermal-chemical (ETC) systems. Instead of computing the gravimetric impetus, it is more useful to study the volumetric energy, which is defined as $I*d/(\gamma - 1)$ [I = impetus, d = density, and γ the ratio of the specific heats at constant pressure and constant volume]. While computing the gravimetric impetus or chemical energy is nowadays a straightforward matter, estimating densities is not. A rough estimate can be obtained by assuming that volumes are additive. In addition, for a given set of ingredients, the composition with the maximum chemical energy turns out to change mildly with the amount of added electricity. Also, the estimated density itself changes with composition. A correct computation of the maximum energy must take both effects into account. In this report, thermochemical requirements for potential ETC propellants which are required to meet weapon performance levels are identified. In addition, a procedure to assess the performance potential of candidate ETC propellants is proposed. This procedure is then applied to several candidate propellants.

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1. INTRODUCTION

The electrothermal-chemical (ETC) propulsion concept is a technology that is believed to offer the potential payoff of increased muzzle kinetic energy (increased velocity or launch mass) within the constraints of current gun envelops (geometric configuration). Figure 1 illustrates the major components of a generic ETC armament system. The propulsion portion of the ballistic cycle is initiated by the discharge of a large electrical current from the power source into the plasma capillary; here, a fuse wire is vaporized to create a high-temperature (10,000–20,000 K) plasma. As electrical current continues flowing through the plasma capillary, the plasma temperature is maintained by ohmic heating, and wall material (usually polyethylene) is ablated because of the high temperatures. The pressure gradient between plasma capillary and combustion chamber forces the plasma to flow into the combustion chamber where it reacts with a propellant, which may be endothermic or exothermic, and generates hot gas, which is the working fluid that accelerates the projectile just as in a conventional solid propellant gun. In theory the electrical energy input to the capillary should determine the nature of plasma discharge in the combustion chamber and the resulting breech pressure. Thus, input of the optimal electrical energy pulse should result in a gun pressure profile that can be tailored to produce maximum performance (i.e., maximum muzzle kinetic energy).

As detailed elsewhere (Wren, Oberle, and Morrison, to be published; Oberle and White 1991; Morrison et al. 1990), utilizing electrical energy to supplement the chemical energy or to supply a major portion of the total energy is an inefficient use of the electrical energy. The major purpose of the electrical energy should be to control the gas generation rate and hence the subsequent pressure history in the gun. Also, to minimize the mass and volume of the electrical power subsystem, the total electrical energy per round must be as small as possible. Therefore, if the potential performance benefits of ETC propulsion are to be realized, the additional energy required for enhanced performance must be predominately chemical. Fortunately, utilizing electrical energy permits the use of a broader range of materials for propellants than in conventional solid propellant systems. The authors believe that propellant materials with the required performance characteristics can be identified.

The overall objective of this report is to indicate goals for propellant energy content and to propose a methodology to aid in the identification of ETC propellants having the desired ballistic characteristics. Specific objectives of this report are:

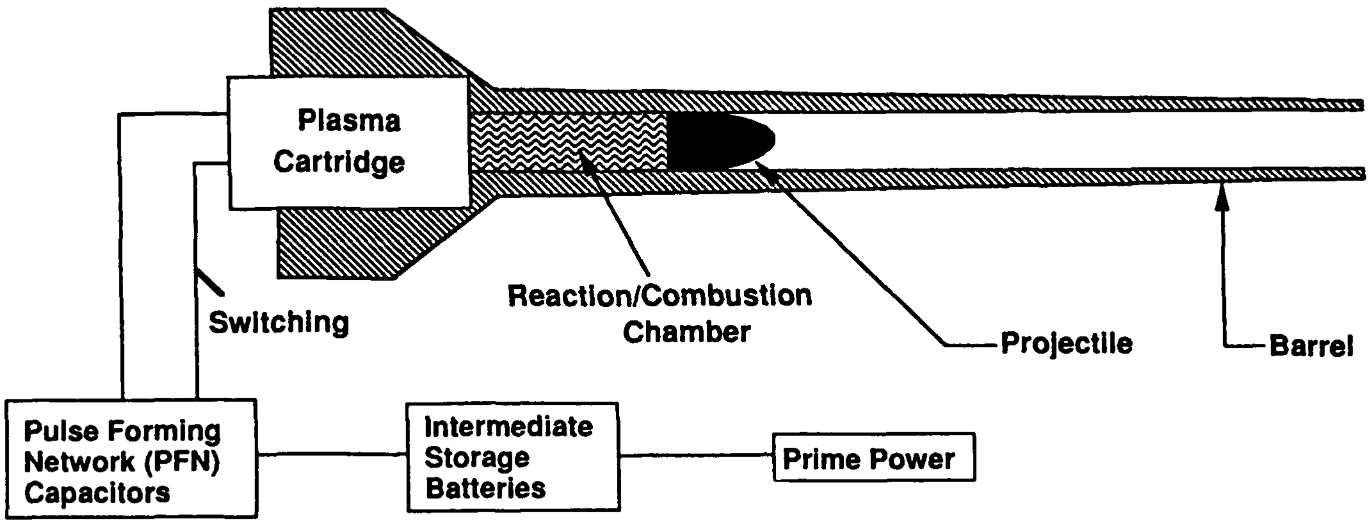

Figure 1. Schematic of Generic ETC Armament System.

(1) provide goals for propellant energy density required to obtain desired ballistic performance;

(2) provide a methodology for the calculation of candidate propellant thermochemical properties; and

(3) illustrate the methodology by the examination of several ETC candidate propellants proposed by the authors.

2 ENERGY DENSITY GOALS

One objective of the Army's ETC propulsion program is to obtain approximately 18 MJ of projectile muzzle kinetic energy in a 120-mm cannon. (Current performance in the M256, 120-mm cannon is approximately 11 MJ. Also, due to the elastic strength profile of the M256 cannon, the maximum muzzle kinetic energy which can be obtained is between 14 and 15 MJ [Oberle and White 1991]. Thus, for this report, calculations are for a generic 120-mm cannon.) In this report, propellant energy density goals will be relative to obtaining this objective. Goals will also be provided for 15 MJ of muzzle kinetic energy, a figure believed by the authors to be the minimum energy of interest in a 120-mm ETC system. First, interior ballistic (IB) calculations using the CONPRESS code (Oberle and White 1991) will be performed.

This code assumes a constant breech pressure to propellant burnout followed by adiabatic expansion. To compensate for energy losses and nonconstant breech pressure profiles measured in actual gun firings, 95% of the computed CONPRESS velocity will be used. Irish (1985) and Morrison (1990) have shown that 95% of the constant breech pressure velocity is a reasonable figure for a well-designed solid propellant gun.

Table 1 provides those inputs for the IB calculations that remain fixed for all calculations. The propellant mass with the chamber volume adjusted for no ullage, the propellant volumetric energy density and the propellant density are parametrically varied. Calculations are performed for a projectile travel of 4.75 m, the current M256 cannon, and for 6.0 m, the proposed travel for a new 120-mm tube.

Table 1. Fixed Parameters for Constant Breech Pressure IB Calculations

Projectile Mass:	11.4 kg
Propellant Thermochemistry:	
Gamma	1.22
Covolume	0.6 cm^3/g
Constant Breech Pressure	574 MPa

Figures 2 and 3 provide potential performance curves for a generic 120-mm bore diameter cannon as measured by muzzle kinetic energy for various charge masses and volumetric energy densities; both cases assume a propellant density of 1.5 g/cm^3. Figure 2 is for a travel of 4.75 m and Figure 3 for a travel of 6.0 m. The assumption of no ullage implies that the loading density (charge mass/chamber volume) is identical to the propellant density, 1.5 g/cm^3. Current high performance solid propellant rounds have loading densities between 0.9 g/cm^3 to 1.0 g/cm^3. Both graphs show that increasing propellant mass does not result in continually increasing muzzle velocity; rather, there is an optimal charge mass for each propellant volumetric energy. Also, from Figure 2, for 4.75 m of travel, to obtain 15 MJ of muzzle energy requires a propellant with a volumetric energy density of approximately 9.5 MJ/L, and 18 MJ of muzzle energy requires well in excess of 12 MJ/L. From Figure 3, the volumetric energies in a cannon with 6.0 m travel are approximately 6.5 MJ/L and 10 MJ/L to obtain 15 and 18 MJ of muzzle energy, respectively.

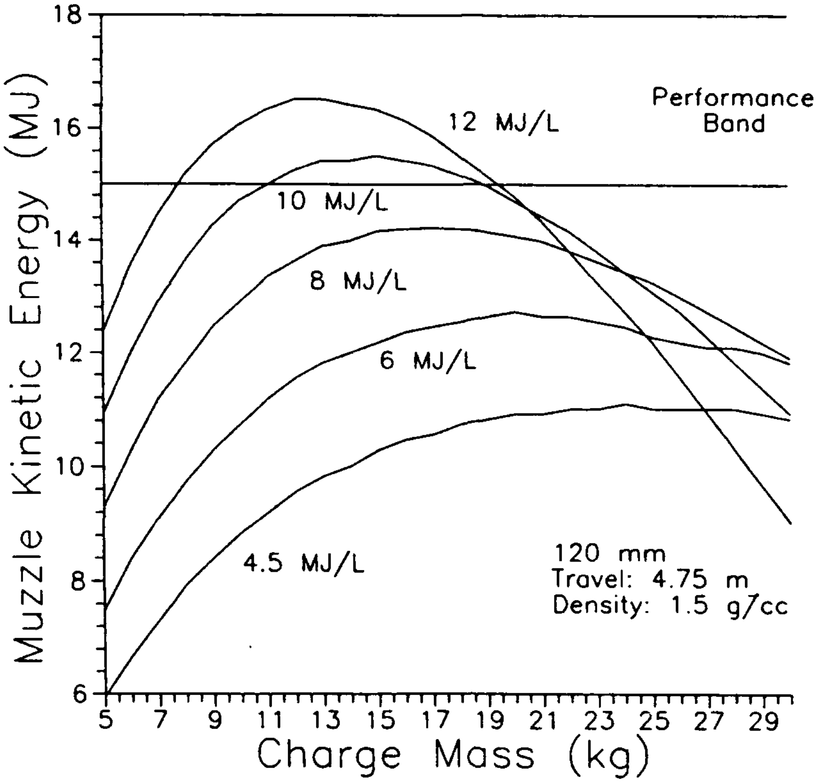

Figure 2. Muzzle Kinetic Energy vs. Charge Mass, 4.75-m Travel for Various Volumetric Energy Densities.

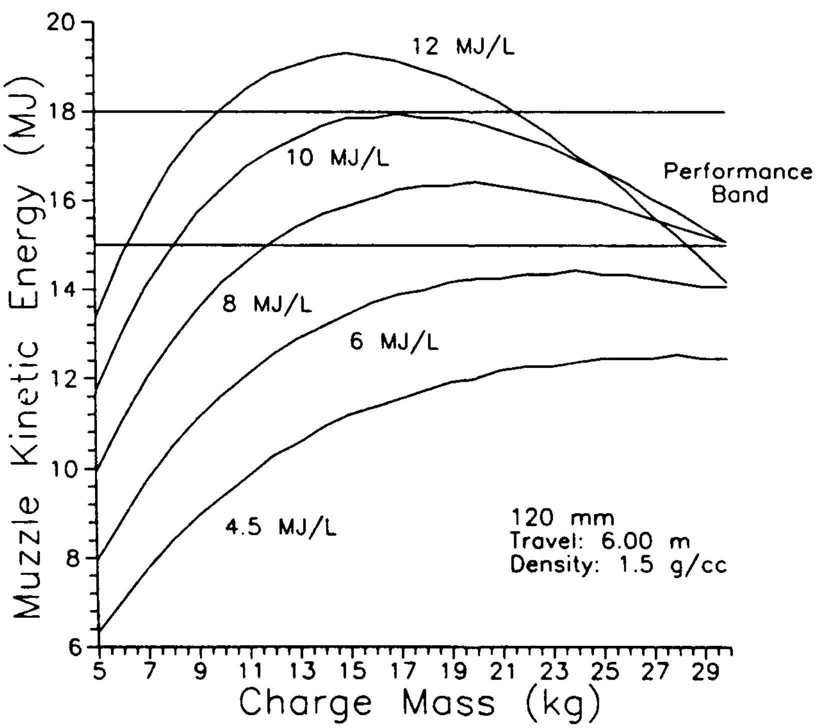

Figure 3. Muzzle Kinetic Energy vs. Charge Mass, 6.0-m Travel for Various Volumetric Energy Densities.

Figures 4 and 5 summarize minimum total energy requirements (chemical + electrical) to obtain 15 and 18 MJ of muzzle energy as a function of the propellant density for projectile travels of 4.75 m and 6.0 m in a 120-mm gun. Volumetric energy (MJ/L) can be obtained by multiplying energy (vertical axis) by propellant density (horizontal axis). The dimensions of energy per unit mass are chosen because propellant energies are generally given gravimetrically, not volumetrically, and it is gravimetric energy that is used in interior ballistic codes. In addition, the gravimetric energy is easily calculated from the results of thermochemical codes by

$$\text{Gravimetric Energy} = \frac{\text{Impetus}}{(\gamma - 1)}. \qquad (1)$$

Figures 4 and 5 can be used to determine both the gravimetric and volumetric energy required to obtain a given muzzle energy as a function of the propellant density. For example, Figure 4 shows that to obtain 18 MJ of muzzle kinetic energy requires a propellant with a combined chemical and electrical energy content of between 11.1 MJ/kg and 10.6 MJ/kg, depending on density. Volumetric energies vary from 11.1 MJ/L to 15.9 MJ/L as the propellant density varies from 1.0 g/cm^3 to 1.5 g/cm^3. For a travel of 6.0 m, Figure 5 indicates that the total energy to obtain 18 MJ of muzzle kinetic energy is between 7.3 MJ/kg and 6.8 MJ/kg, depending on density. Volumetric energies range from 7.3 MJ/L with a propellant density of 1.0 g/cm^3 to 10.2 MJ/L with a propellant density of 1.5 g/cm^3. Although the volumetric energy density required to obtain a given muzzle energy increases as the density increases, this does not imply that low density propellants are necessarily better than high density propellants. For a given performance level, approximately the same amount of total energy will be required regardless of the propellant density. Then, the higher the density the less volume required for the propellant, which will have a positive impact on breech design. Although both the chemical and electrical energies are included, practical applications will limit the electrical energy input to about 0.5 MJ/kg of propellant. Thus, the propellant energy requirements will not be drastically reduced.

In summary, the goal required for candidate ETC propellants to produce 18 MJ of muzzle kinetic energy in a 120-mm cannon is a propellant with a volumetric energy density of at least 11.1 MJ/L if projectile travel is 4.75 m. Use of an extended travel tube lowers the volumetric energy density requirement by about 34% (11.1 MJ/L for 4.75-m travel to 7.3 MJ/L for 6.0-m travel). The actual value for a specific propellant will depend upon the propellant density as shown above. Furthermore, these

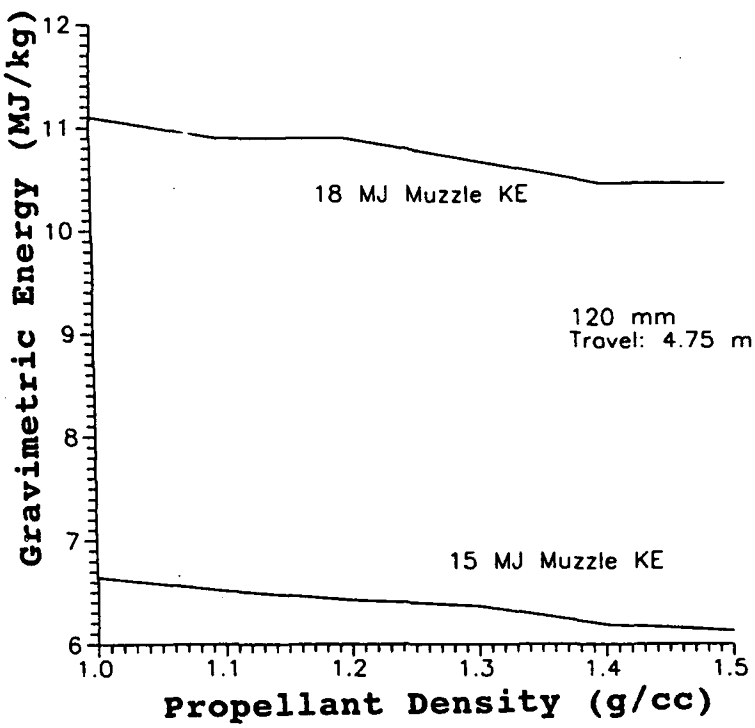

Figure 4. Required Energy vs. Propellant Density for Performance Levels of 15 and 18 MJ of Muzzle Energy, Travel = 4.75 m.

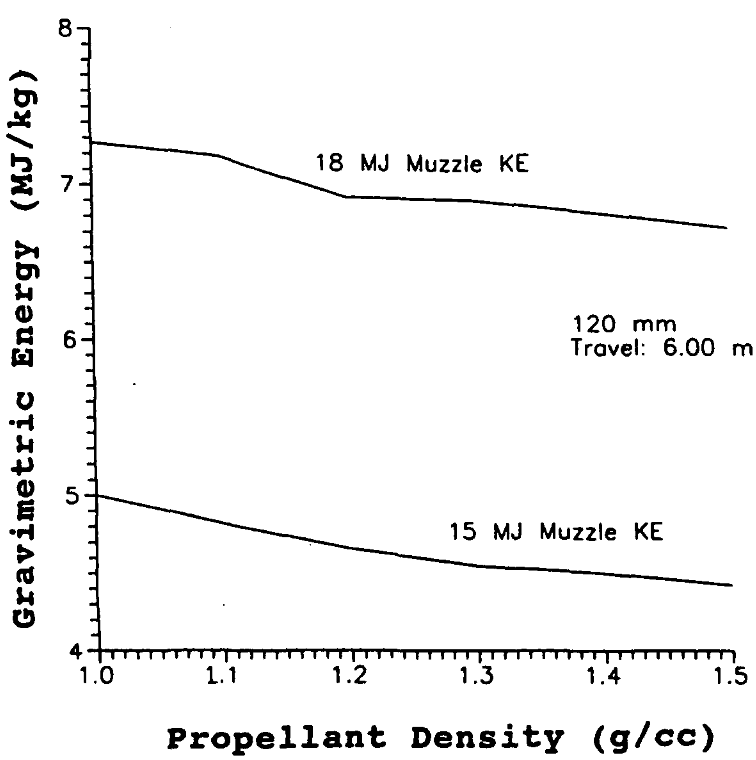

Figure 5. Required Energy vs. Propellant Density for Performance Levels of 15 and 18 MJ of Muzzle Energy, Travel = 6.0 m.

values are based on the assumption that propellant loading density is the same as the propellant density. Inclusion of plasma capillaries or extender tubes or the requirement of ullage in the breech will generally increase the volumetric energy density requirement.

3. PROPELLANT IDENTIFICATION METHODOLOGY

3.1 Outline of the Method. The method proposed by the authors comprises five steps:

(1) Selecting a figure of merit for ranking proposed compositions.

(2) Selecting a procedure for estimating the density of propellant mixtures.

(3) Assembling a set of programs that will compute the impetus of the propellant mix, estimate the density of the starting composition, and produce a table containing estimated density and volumetric ballistic energy as a function of composition.

(4) Restricting the search.

(5) Evaluating candidate propellants; seeking compositions with maximum volumetric energies (if these maxima exist).

3.2 Details.

3.2.1 The Figure of Merit. The figure of merit to be chosen must be appropriate to volume-limited systems. There are two candidates. The first is the volumetric impetus, I_V, defined as $I_G * \rho$, where I_G is the familiar gravimetric impetus. The other is E_V, which in this report will be termed the volumetric ballistic energy, defined as

$$E_V = \frac{I_G * \rho}{\gamma - 1} \quad , \tag{2}$$

where γ is the frozen gamma (ratio of the two specific heats). For this report, E_V is chosen as the figure of merit. Although this choice is debatable, it was selected for several reasons. First is the fact that

energy, not impetus, is the important quantity in ballistic performance codes. Next, the volumetric energy takes into account the effect of propellant density, which may vary over a wide range for proposed ETC propellants. Finally, this value can be computed based solely on propellant composition and thermochemistry. As long as the density and ratio of specific heats stay within the ranges 1.0 g/cm^3 to 1.8 g/cm^3 and 1.1 to 1.3, respectively, and only a small amount of solid particulates are formed in the combustion products (White and Oberle 1992), the authors believe that the volumetric energy density is an appropriate choice for a figure of merit. As will be seen later, this decision affects the selection of the "best" composition.

3.2.2 Estimating the Final Density. The impetus and, thus, energy of any propellant is readily computed by various thermodynamic programs; density, however, is not so readily obtained. The estimation of the density, ρ_f, of a solution of two or more ingredients from only a knowledge of the densities of its components is no easy task. In the present case, however, it appears that almost all of the candidate systems will be composed of components that are immiscible with each other. In such cases, there is no interaction between the components (before combustion), and so their volumes are additive.

Consider a system of volume V and mass W containing w'(1) and w'(2) grams of immiscible ingredients 1 and 2; w'(1)/W = w(1) and w'(2)/W = w(2) are the weight fractions of the ingredients.

The additivity assumption means that the final volume of this system is given by

$$V = [w(1)v(1) + w(2)v(2)] * W \quad , \tag{3}$$

where v(i) is the specific volume of ingredient i; hence, the density of ingredient i, $\rho(i)$, is $\rho(i) = 1/v(i)$.

Equation 3 can therefore be written as

$$V = \left[\frac{w(1)}{\rho(1)} + \frac{w(2)}{\rho(2)}\right] W \quad . \tag{4}$$

Since

$$1/\rho_f = V/W \quad , \tag{5}$$

substitution of Equation 4 into 5 produces

$$\frac{1}{\rho_f} = \frac{w(1)}{\rho(1)} + \frac{w(2)}{\rho(2)} \quad , \tag{6}$$

or

$$\rho_f = \frac{\rho(1)\rho(2)}{w(1)\rho(2) + w(2)\rho(1)} \quad . \tag{7}$$

It is readily shown that Equation 3 is exactly equivalent to assuming that the molar volume of the system is the sum of the mole-fraction weighted sum of the molar volumes of the components.

3.2.3 Computational Procedure. All of the thermodynamic computations are performed with a modified version of the BLAKE code (Freedman 1982) run at a loading density of 0.2 g/cm^3. In addition to its usual output, this version also produced a summary file containing the title of the run and the usual summary of the propellant gases' thermodynamic properties. This summary file is in turn used as input to a post-processing program that combines composition and thermodynamic data to produce an estimate of the composition's density and the volumetric impetus and energy. The output from this program is two files—one labelled and formatted for reading and one that was for input to a graphics program.

3.2.4 Restricting the Search. Since not more than 0.5 MJ/kg of electrical energy will be available, only exothermic propellant formulations need be considered. Fortunately, this still leaves a wide field.

• Mono- or Bipropellant? From the first it appears unlikely that any conventional monopropellant can meet the requirements. For example, JA2, an energetic solid propellant, has a gravimetric ballistic energy of 5.6 MJ/kg when augmented with 0.5 MJ/kg of electrical energy. If the gun loading density equals the material density, 1.6 g/cm^3, the resulting volumetric energy is 8.96 MJ/L, but this assumption is not realistic. A more realistic maximum gun loading density is 1.2 g/cm^3; this leads to a value of 6.72 MJ/L, which is below the target values of approximately 7 and 11 MJ/L.

The energy of JA2 or similar propellants can be increased by increasing their oxidizer content. Practically, this means adding more nitroglycerin or diethyleneglycol dinitrate, which would in turn increase both hazard potential and flame temperature. However, if instead of mixing the oxidizer with

the fuel, it is possible to keep them separate until the moment of combustion, potential hazards could be reduced. Unfortunately, neither nitroglycerin or diethyleneglycol dinitrate are attractive choices for the oxidizer because of their hazards and adverse physiological properties (Meyer 1987).

Accordingly, attention is focused on finding energetic bipropellant combinations of oxidizers and fuels with high densities.

- Choice of Oxidizer. One choice for the oxidizer is a solution of 81.1% hydroxylammonium nitrate (HAN) in water (which is a 13-molar solution). This mixture has a density of 1.54 g/cm^3 (Sassé et al. 1988) which is attractive, and its safety and toxicity have been intensively studied in recent years. For convenience, this solution will be referred to simply as HAN in the remainder of this report.

- Choice of Fuel. Here, too, high density (1.5 g/cm^3 or greater) is a necessary but not a sufficient condition—promising compounds must also be intrinsically energetic.

Densities above 2 g/cm^3 are more often found in metals or inorganic compounds, but these compounds are not promising fuel ingredients. The principal reason is their generally low heats of combustion. There are some exceptions; e.g., hydrides. Thus, titanium hydride (density = 3.75 g/cm^3) has been briefly considered. A mixture of 24% TiH_2 + 76% HAN/H_2O (which is not optimal) with 0.5 MJ/kg of added electrical energy has a volumetric energy of 8.17 MJ/L. Even if this value were large enough, there is the unsettled question of whether the large amount of solid product in the exhaust gases (about 23% Ti_2O_3) would interfere with the proper operation of the gun (White and Oberle 1992). Such compounds are not considered further in this report.

Organic compounds containing only C, H, N, and O almost always have densities less than 1.3 g/cm^3. There are some interesting exceptions; e.g., 7-methyl uric acid has a density of 1.706 g/cm^3, but it is not energetic. The principal exceptions are nitrates and nitro compounds.

3.2.5 Application. Application of the screening methodology is provided for several candidate propellant formulations in the next section.

4. EXAMPLES

No systematic search of the literature is undertaken; the present work is restricted to considering some promising compounds from Meyer (1987). The performance level that a suitable ETC system must reach is high; it is not immediately obvious that satisfactory propellants can be found that will achieve them. Therefore, in investigating various possibilities, very little effort has been made at this time to screen them for safety, toxicity, producibility, or other requirements that all propellants must meet before being fielded. Additional details can be obtained in a report by Boggs et al. (1991).

4.1 Selected Fuels. Three fuels of interest are shown in Table 2.

Table 2. Three Illustrative Fuel Components[a]

Chemical Name	Abbreviation	Density (g/cm^3)	Formation Enthalpy (kcal/mol)	Ballistic Energy[b] (MJ/L)
Cyclo-1,3,4-trimethylene-2,4,6-trinitramine	RDX	1.82	14.69	11.00
Trinitro-2,4,6-phenylmethylnitramine	tetryl	1.73	8.08	8.00
Trinitroaniline	TNA	1.76	-13.66	6.18

[a] All data are from Meyer (1987).
[b] Computed from BLAKE thermodynamic values.

4.2 Variation With Composition or Added Energy. Earlier in this report, it was stated that the volumetric energy density was selected as a figure of merit instead of the volumetric impetus; several reasons for this choice were given. In this section, the rationale for selecting the volumetric energy density over the volumetric impetus will be further illustrated as the composition dependence of the results are explored.

All three fuels are more energetic than 81.1% HAN (volumetric energy = 1.18 MJ/L). Consequently both gravimetric and volumetric impetuses, considered as a function of composition, increase to the maximum value of the pure fuel. Figure 6 illustrates this result for the gravimetric and volumetric impetuses of HAN/tetryl and HAN/TNA. Figure 7 shows the composition dependence of the gravimetric

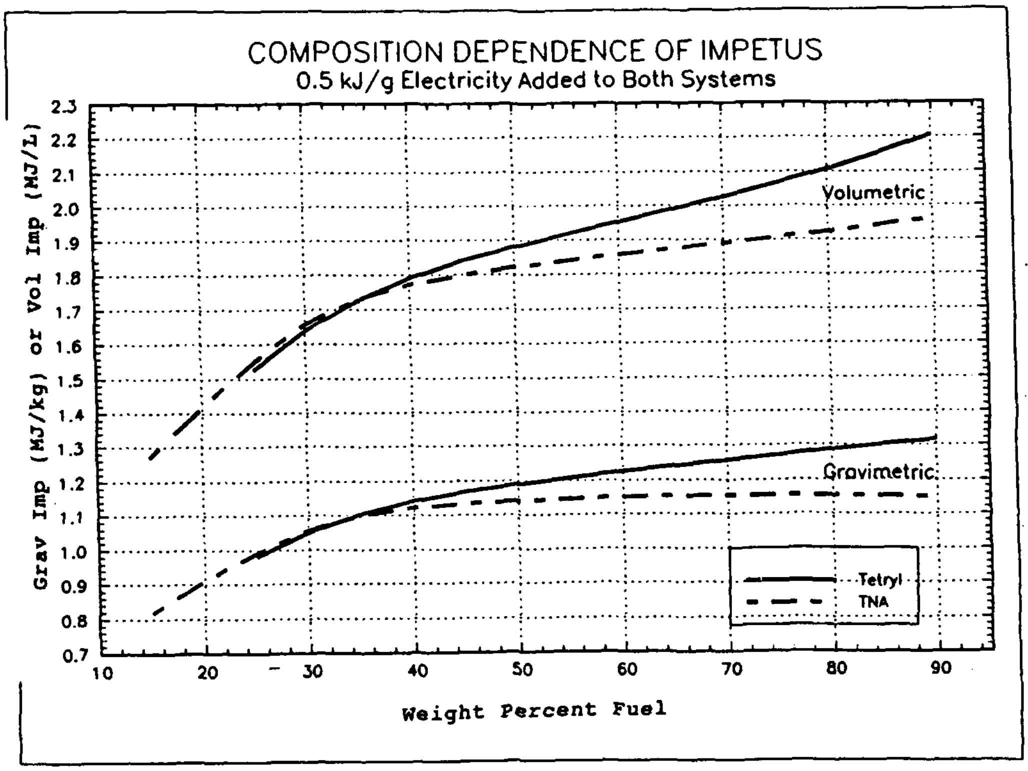

Figure 6. Gravimetric and Volumetric Impetuses of HAN/Tetryl and HAN/TNA, 0.5 MJ/kg Electrical Energy Added.

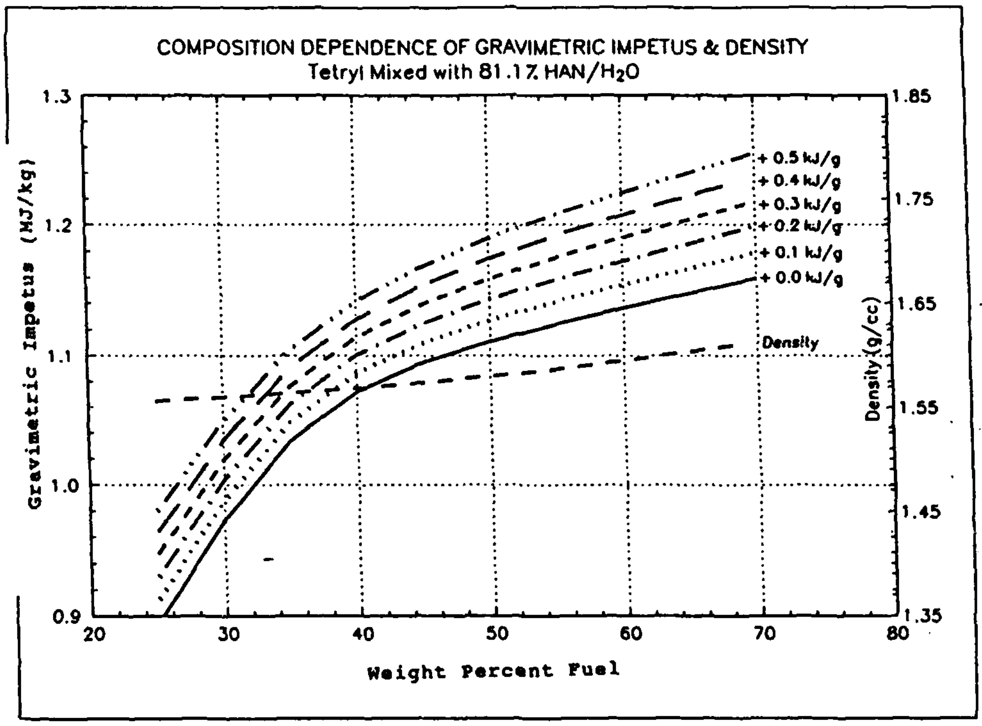

Figure 7. Gravimetric Impetus and Density vs. Percentage of Fuel, Tetryl Mixed With 81.1% HAN/H_2O.

impetus of HAN/tetryl for six different levels of added electrical energy. In all of these cases, the impetus rises monotonically to the value of the pure fuel. Figure 7 also shows the composition dependence of the estimated density, which is virtually linear. Thus, the dependence on composition of volumetric impetus (density * gravimetric impetus) will exhibit the same behavior as the gravimetric impetus.

The volumetric and gravimetric energies, however, sometimes behave differently. Figure 8 shows the composition dependence of the volumetric energy of these same two systems. Perhaps surprisingly, there is now a maximum. The reason for this is the concentration dependence of gamma, which is shown in Figure 9 for the HAN/tetryl mixture. It has a pronounced minimum, so that $1/(\gamma - 1)$, in turn, has a maximum, which produces maxima in the gravimetric and volumetric energies. In general, these maxima occur at different concentrations.

Figure 10 shows the volumetric energy for the same system on a larger scale. The maximum is quite evident. The location of this maximum occurs at higher fuel concentrations as the electrical energy increases. This point is emphasized by the sloping line drawn through the maxima.

Since the effective electrical energy consumed per gram varies throughout the discharge cycle, this dependence of the maximum total energy on the amount of added electricity means that, theoretically, there cannot be a truly optimum mixture in this system. This may have implications for charge design. However, Figure 10 indicates the actual shift is not large and should not constitute a problem.

Such a maximum in the volumetric (gravimetric) energy does not occur in every HAN-fuel system; for example, Figure 11 shows that it does not occur in HAN-RDX. The explanation is the same: the change (or lack of it) in the gamma of the product gases with the initial propellant composition.

5. ADDITIONAL DISCUSSION

5.1 <u>The System HAN + RDX</u>. Perhaps the most interesting system of the four considered here is 81.1% HAN and RDX. The volumetric energy of this system increases up to neat RDX as the weight-percent of fuel approaches 100%. For a weight-percent of fuel above 65%, the system with 0.5 MJ/kg of added electrical energy has a volumetric energy greater than 10 MJ/L. This suggests the possibility of a fuel composed of RDX and a high-density binder. Another possibility is based on the well-known

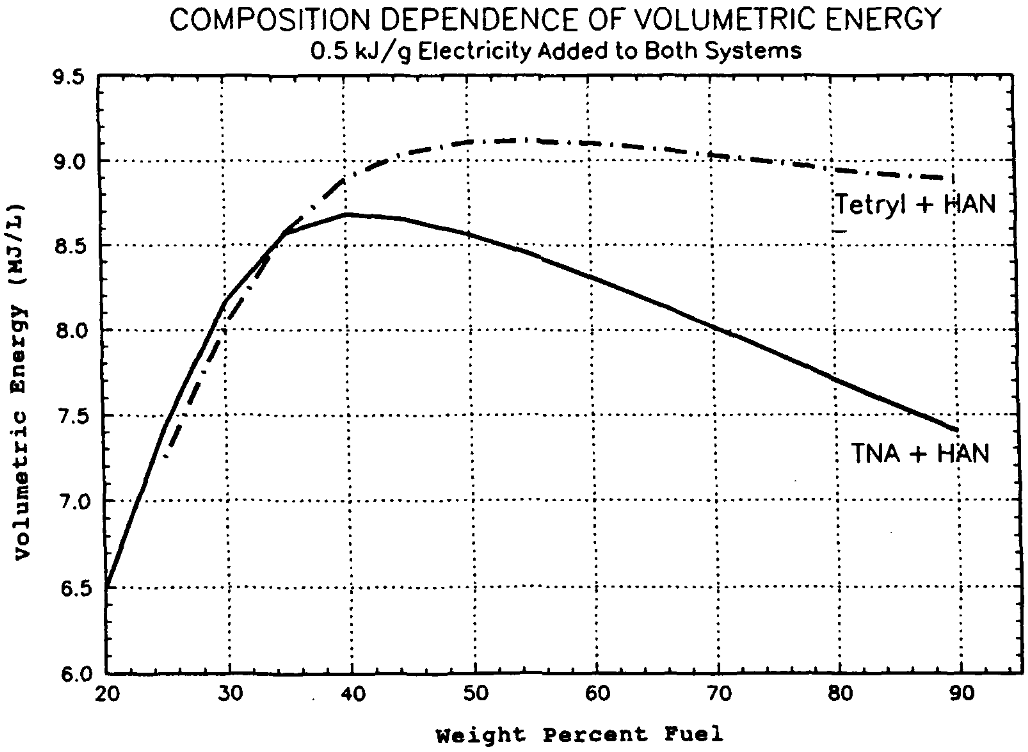

Figure 8. Volumetric Energy vs. Percentage of Fuel, Tetryl + HAN, TNA + HAN, 0.5 MJ/kg Added Electrical Energy.

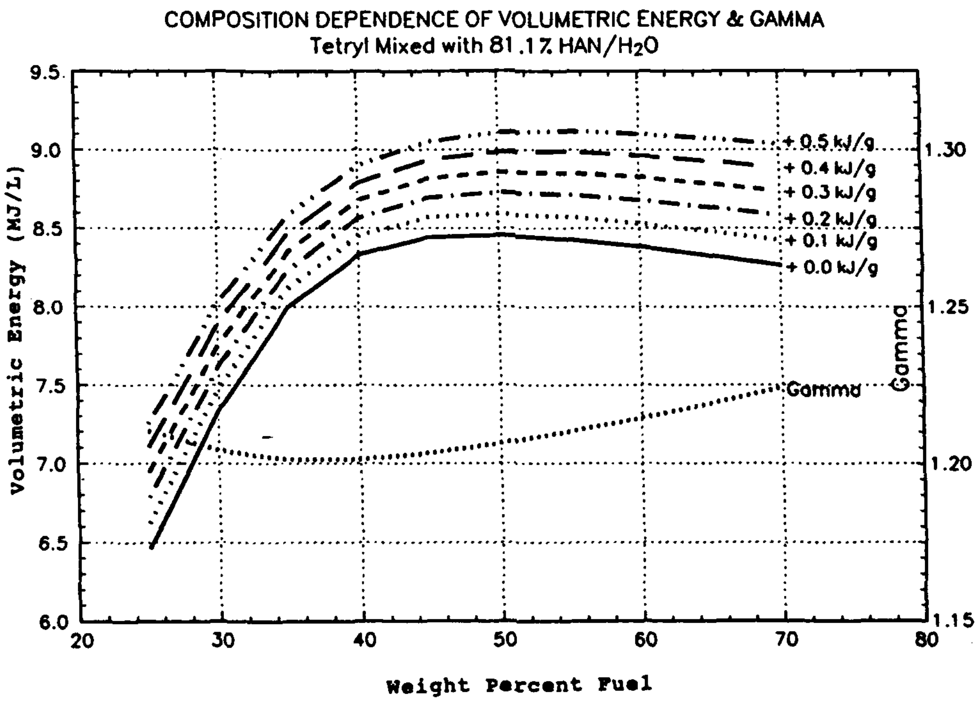

Figure 9. Volumetric Energy and Gamma vs. Percentage of Fuel, Tetryl Mixed With 81.1% HAN/H_2O.

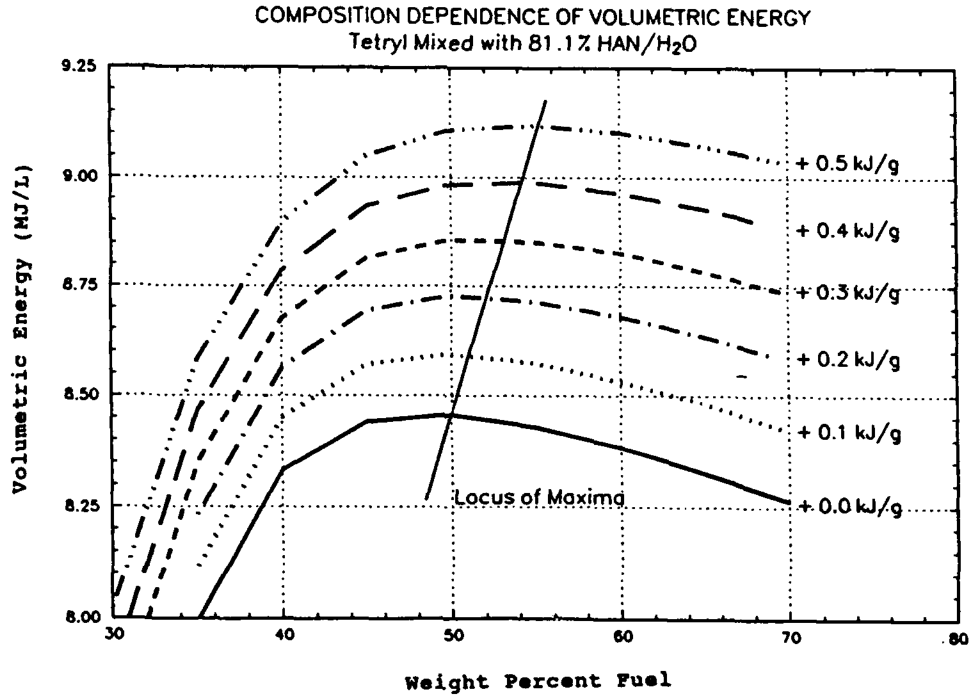

Figure 10. Volumetric Energy vs. Percentage of Fuel, Tetryl Mixed With 81.1% HAN/H_2O (Figure 9 Expanded).

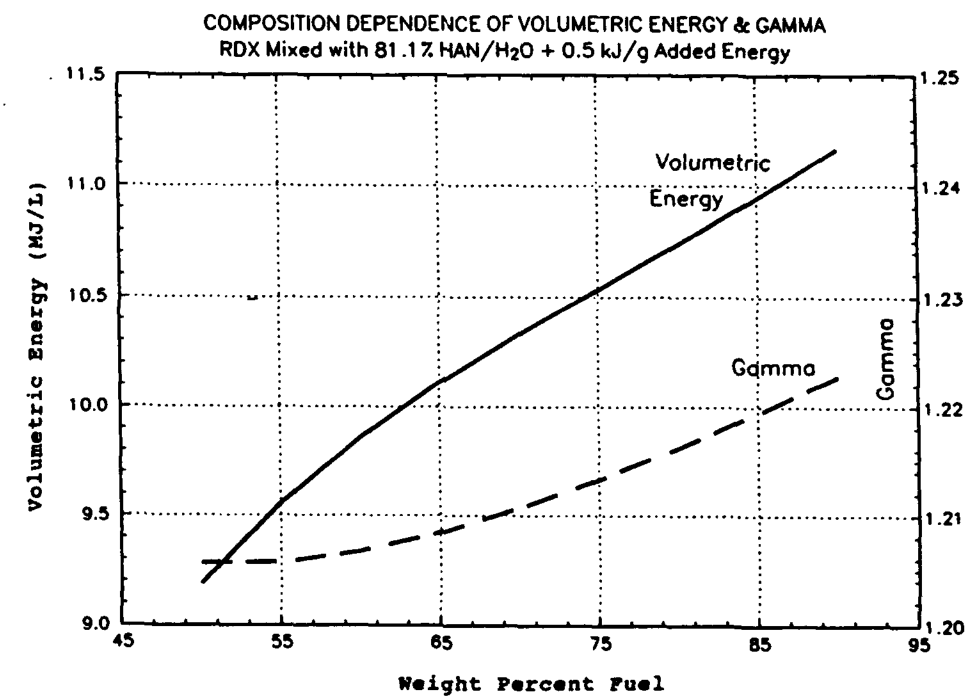

Figure 11. Volumetric Energy and Gamma vs. Percentage of Fuel, RDX Mixed With 81.1% HAN/H_2O, 0.5 MJ/kg Added Electrical Energy.

insensitivity of triamino-trinitrobenzene (the so-called "wooden explosive"), so that mixtures of it with RDX may have promise as an ETC fuel component.

5.2 Flame Temperatures. The systems discussed here have been chosen primarily for illustrative purposes. Nevertheless, we must point out a potential drawback to some of them—they have flame temperatures greater than that of JA2 (3,430 K), which has been suggested as an upper limit (Wren and Oberle 1991).

Table 3 shows the properties, including the flame temperatures, for the systems that have been discussed here.

Table 3. Thermodynamic Properties of Selected HAN-Fuel ETC Systems
0.5 MJ/kg of Electricity Added in All Cases

Name	Fuel (%)	Flame Temp (K)	Grav Imp (J/g)	Co-Vol (cm^3/g)	Gamma	Estd Dens (g/cm^3)	Volumetric Energy (MJ/L)
RDX	70	3,912	1,308.9	0.949	1.2107	1.662	10.3
"	75	3,981	1,337.5	0.966	1.2133	1.680	10.5
"	80	4,047	1,365.5	0.983	1.2162	1.700	10.7
"	85	4,110	1,393.5	0.999	1.2194	1.723	10.9
"	90	4,173	1,421.6	1.015	1.2228	1.750	11.2
Tetryl	54	3,666	1,204.4	0.937	1.2094	1.586	9.1[b]
TNA	41	3,423	1,124.2	0.886	1.2047	1.582	8.7[b]
TATNE[a]	38	3,362	1,092.1	0.868	1.2063	1.595	8.4[b]

[a] Triamino-trinitrobenzene

[b] Maximum value for this system

6. SUMMARY

The goal for candidate ETC propellants should be to identify a propellant with a volumetric energy of at least 11 MJ/L in order to obtain 18 MJ of muzzle kinetic energy in a generic 120-mm cannon.

Volumetric energy is the most appropriate figure of merit for comparing candidate propellants due to the wide range of proposed ETC propellant densities and the effect of the ratio of specific heats. The density of mixtures of immiscible components can be estimated by assuming volume additivity. Useful ETC propellants will have to be both energetic and have a high density. A concentrated (13-molar) solution of HAN is a promising oxidizer for such propellants. Possible high-density fuels include nitro compounds, some of which have already been characterized as explosives. The volumetric impetus of systems of HAN plus more energetic fuels increases monotonically with fuel concentration; but the volumetric and gravimetric ballistic energy of some of these systems exhibit a maximum due to the composition dependence of the ratio of specific heats. Fuels containing RDX and a less-energetic fuel or a high-density binder deserve further consideration.

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